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Scenarios for Nuclear Fusion in Palladium-Deuterium Alloys at Ambient Temperature

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Abstract: The quantum aspects of exothermic reactions in palladium-deuterium alloys possibly also containing lithium and/or exotic systems $X^{\ominus}$ or $X^{\oplus}$ (discussed by *Glashow, Cahn,* and the writer in 1981 [12, 13]) are highlighted.

In addition to various announcements in newspapers and at press conferences, an eight-page paper «Electrochemically Induced Nuclear Fusion of Deuterium» was published on April 10th, 1989 by *Fleischmann* and *Pons* [1]. This discovery – if confirmed – has many of the aspects of the entirely unexpected, the dimly understood, and contrast to the mainstream of contemporary thought (which characterized [2] the discovery of X-rays by *Röntgen* in 1895 and of radioactivity by *Becquerel* in 1896) and was here analyzed by my colleague *Augustynski* [3]. We decided to share the work by me writing a shorter comment on the conceivable quantum physics behind the reported observations. One should not confuse the obvious difficulties of quantitative numerical treatment [4, 5] of quantum chemistry involving elements having two-digit Z values with more direct conceptual corollaries of quantum mechanics. The palladium alloys may present unique quantum-mechanical opportunities of isotopes of hydrogen (or higher atomic numbers Z) transgressing the ideas prevailing among chemists [5] since about 1935 that condensed matter consists of «matter-of-fact» nuclei (characterized by quantum numbers [2] Z and A) embedded (at practically fixed positions) in a delocalized density distribution of far lighter electrons.

It may be worthwhile noting that palladium forms entirely different types of alloys with the three univalent elements hydrogen, lithium, and silver. The cubic closed-packed $Pd_{1-x}Ag_x$ is fully miscible; the alloys $Pd_{1-x}Li_x$ allow most x values but are only isotypic with pure Pd up to $x \approx 0.06$, and comprise distinct types like $LiPd_7$ and two types (LiRh and CsCl) close to LiPd [6, 7]. The binary alloys PdH_x are interstitial (and not substitutional) and have been extensively studied [8]. Compositions between $x = 0.03$ and 0.6 are mixtures of two isotypic phases (like the high- and low-density cubic modifications of metallic cerium), the α-phase expanding the unit cell a_0 at most by 0.1 percent going from $x = 0$ to 0.03, and the β-phase starting at $PdH_{0.61}$ (or $PdD_{0.55}$) having $a_0 = 4.026$ Å increased 3.5 percent relative to Pd, and hence the volume per Pd 11 percent higher (differential mole volume of H only 2 mL). Under electrochemical conditions far from thermodynamical equilibrium [1, 3] x can approach 1 or even show higher values. There is one specific feature in common for the H, Li, and Ag alloys: The magnetic susceptibility decreases linearly [8] with x in PdH_x (and PdD_x) from a moderate positive value to diamagnetism a little beyond the β-phase limit, close to $PdH_{0.66}$ (this is less unexpected, because of the coexistence of the two cubic phases) the same way as from Pd to isotypic $Pd_{0.4}Ag_{0.6}$; and also [7] from Pd to $Pd_{0.75}Li_{0.25}$ (but less regularly linear, concomitant with the many intermediate alloy phases).

We return below to opinions about H positions in PdH_x in the discussion of three plausible scenarios, but would like to de-emphasize earlier concepts of H being present as atoms *or* protons *or* closed-shell $H^{\ominus}$. The carrier of the hydrogen characteristics *is* the proton (also in NH_3 or CH_4) and its influence on the adjacent electron density is intricate, and smoothly varying. By the same token as hydrogen atoms cannot be recognized in $PdH_{0.6}$, *atoms* of silver, palladium (in the vapour around 3000 °C really being noble-gas atoms like mercury vapour) and lithium have no relevance for the description of the metallic elements and alloys.

First Scenario: Two Deuterons Colliding

If there are definite interstitial sites available for nuclei having $Z = 1$, the most obvious choice is the «octahedral sites» in cubic close-packed elements, six Pd nuclei being situated on Cartesian axes at a distance $a_0/2 = 1.95$ Å from the origin available to the proton. Since each unit cell of volume a_0^3 contains four Pd and four such H sites, the lower (and most stable) β-phase $PdH_{0.66}$ would show two-thirds occupation. The need for oversaturated hydrogen alloys [1, 3] might be justified by the necessity of two adjacent deuterons on the same site, being a constitutional feature for $x > 1$. The opposite approach of standing De Broglie waves (suggested below) would not increase the rate by much more than x^2. It may also be asked how specific the octahedral H site is for a small, core-less entity like a deuteron. The distance on a straight line connecting two Pd sites is still 1.37 Å, perhaps permitting a mildly fluctuating deuteron density in a «Swiss cheese» distribution of sites, having the holes filled with Pd cores.

Dramatic variations of the Coulomb potential close to nuclei do not induce strong singularities in wave-functions, as seen from the almost constant amplitude of the 1s ground state of a hydrogen atom passing the nucleus (large modifications of the amplitude would increase the local kinetic operator far more). Though the interaction of two deuterons is repelling (and not attracting) they would still meet with a constant (but very small) amplitude. The cross-section of such encounters is enhanced by low velocity v determining the De Broglie wave-length $\lambda = h/mv$. At a given T, the average v of deuterons is 0.71 times that of neutrons, and λ of the order of 1 Å at 50 °C. Before comparing with the well-known behaviour of thermalized neutrons as a gas, we may add that (anisotropic) lower v and longer λ may occur in a crystal for positive particles. The cross-section for absorption of neutrons in a given isotope is far below $\lambda^2 = 2 \cdot 10^{-16}\,cm^2 = 200$ million barn. The highest value known [9] for a radioactive isotope is 2.64 million barn for ^{135}Xe, and for a stable isotope 0.25 million barn for ^{157}Gd. It is between 20000 and 60000 barn for ^{113}Cd, ^{149}Sm, and ^{155}Gd. These high values for some elements having three-digit A values are related to extremely sharp peaks in the neutron absorption curve as a function of neutron kinetic energy. Light elements tend to have cross-sections close to the «target area» of the nucleus being a few barn; and neutron cross-sections below 10^{-4} barn [9] are known (also for ^{209}Bi) and it is strictly zero for ^4He. Hence, it is not easy

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to predict what the deuteron-deuteron cross-section will be for small v (it is likely to increase at *lower T*) but it can easily be 10^{-20} barn, a given deuteron (confined in the alloy) passing roughly 10^{12} other deuterons every second, having a probability of order 10^{-21} of coalescing each time. The rigid solid alloy may enhance the effective cross-section by phenomena analogous to the recoil-less resonance of Mössbauer nuclei. Exothermic effects above 400 eV/mol D (2 g deuterium in 10 cm^3 alloy) were reported to occur[1] in 10^6 s time, corresponding to 0.02 percent of the ^2H undergoing the reaction to $^3H + {}^1H$, or a relative rate of $2 \cdot 10^{-10} s^{-1}$.

In stellar interiors, the two sets of products ($^3H + {}^1H$) and ($^3He + {}^1n$) have comparable probabilities of formation, but one of the more enigmatic sides of the palladium alloy[1] is that the number of neutrons detected was many million times too low, relative to expectations from the exothermic effect. In the scenario of low-T De Broglie waves, the reaction $^2H + {}^2H$ may have two specific advantages: one is the boson character of ^2H (in contrast to ^1n, ^1H, and ^3H) which does not need local antisymmetrization (keeping two fermions apart), and another that a deuteron is a loose, extended system (compared with usual nuclear densities) and the two positive charges may remain comparatively distant a given instant, but still allowing a significant probability of transferring a neutron from one D ($\equiv {}^2H$) to the other. One may ask why two protons do not absorb an electron in the alloy (and emit a neutrino) forming ^2H. However, even at kT around 1000 eV prevailing in stellar centres, this process has an exceedingly low rate of order $10^{-17} s^{-1}$ whereas D is known to react far more rapidly.

Second Scenario: Lithium Infiltration

As said by *Fleischmann* at lectures in Lausanne (EPFL) and in Genève (CERN)[3], the solution of $Li^{\oplus}OD^{\ominus}$ in D_2O surrounding the palladium electrode[1] cannot be replaced by $Na^{\oplus}OD^{\ominus}$. It is possible[3] that $Li^{\oplus}$ may penetrate the Helmholtz double-layer and occur in small concentrations inside the metal. The affinity of lithium to palladium is very large, and the deuterium oversaturated metal is fairly reducing. *If* the exothermic reaction was $^6Li + {}^2H \rightarrow {}^4He + {}^4He$ (5.8 times more exothermic than $^2H + {}^2H$) the anomalous low number of neutrons detected could be rationalized by this collision of two bosons (if very few neutrons originate from $^7Li + {}^2H$).

Ternary alloys[10] such as $LiPtH_{0.66}$ and $LiPdH_{0.82}$ have been studied. In the latter alloy, hydrogen stays on sites with four Pd and two Li close neighbours, and does not occupy the other sites with four Li and two Pd. One is advised[11] to be cautious, if attempting to prepare $LiPdD_{0.82}$ or other Li_yPdD_x.

A related study[11] involved positive muons (lifetime a few microseconds) in β-phase $PdH_{0.74}$. At $-253\,^{\circ}C$, the muon stays in the middle of 12 sites occupied by (highly mobile?) protons. At higher T, the system rearranges in a way having fewer nearest neighbour protons.

One might argue that the two strongly bound (60 eV) 1s electrons of $Li^{\oplus}$ dramatically decrease the probability for ^2H interacting with the boson ^6Li. This may be true, but it is not so obvious, seen from the point of view of De Broglie waves, since the rate may be that of an inverted tunnel effect.

Third Scenario: Exotic Catalysts

Already in 1981, it was discussed[12,13] whether heavy, long-lived (protected by selection rules of the same kind as the baryon conservation) particles or systems $X^{\ominus}$ might form strong adducts with conventional Z-nuclei, behaving chemically as heavy ($Z - 1$) isotopes. This is particularly significant for the lithium isotope X^8Be not presenting the bottleneck (^8Be instantaneously splits to two ^4He) of stellar nucleosynthesis. Today[2], such $X^{\ominus}$ are called WIMP (= Weakly Interacting Massive Particles), and invoked by many astrophysicists to render stellar interiors easier to rationalize. Jupiter (and it seems also Saturn and Uranus) irradiate more than twice as much heat than absorbed from solar radiation, and low-intensity underground $X^{\ominus}$ nuclear reactions are suspected[12].

In practice, available metallic palladium has undergone solution chemistry and is not too likely to contain $X^{\ominus}$ or $X^{\oplus}$ adducts (as may occur[14] in goldnuggets and native copper). However, deuterium and lithium compounds may (sometimes?) contain trace levels of such species, catalyzing the novel exothermic reaction efficiently.

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